

Application No.: 09/695,232

Docket No.: 21776-00055-US

**REMARKS**

Reconsideration of claims 1-8, 10-14, 16, 17, 20, 22-37 and 39-42 is respectfully requested. Claims 24 and 26 are amended as suggested by the examiner.

The examiner denies patentability of claims 1-8, 10-14, 16, 17, 19, 20, 22-37 and 39-42 under 35 USC 102(b) citing the following four Patent Specifications: (Cited Reference 1) EP 0732139 A2; (Cited Reference 2) EP 0592809 A1; (Cited Reference 3) WO 00/48717; and (Cited Reference 4) USP 5439706. Applicant traverses the 102(b) rejection for each of the cited references.

For reasons described below, each of the four cited references fail to anticipate the applicants' invention. In particular, none of the four cited references teach that by including small amounts of Nb and/or Ta in the oxide, the perovskite type structure is stabilized, thereby leading to the improved characteristics of applicants' claimed materials.

1. Cited Reference 1 [EP 0732139 A2]

EP 0732139 A2 (the '139 application) discloses a tubular solid-state membrane module for isolating oxygen from oxygen-containing gas. The compositions of mixed conducting oxides described in claims 11 to 14, and on page 4, lines 14 to 24 of the specification of the '139 application, form a tubular dense mixed conducting oxide layer or a tubular channel-free porous support. The disclosed composition range of claim 11 is extremely broad, being expressed by the formula,  $A_x A'_{x'} A''_{x''} B_y B'_{y'} B''_{y''} O_{3-z}$ . The disclosed composition encompasses several thousands, if not tens of thousands, different compounds. A preferred range of the composition is described in claims 12 to 14. The elements A, A' or A'' preferably comprises Group 2 metals selected from Ca, Sr, Ba and Mg. A more preferred composition is defined as  $La_x A'_{x'} Co_y Fe_y Cu_{y''} O_{3-z}$ .

In contrast, applicants' mixed conducting oxide of formula (1) as described in claims 1 and 2 of the present application define a composition range of a mixed conducting oxide in perovskite structure containing Fe or Co, and at the same time Nb or Ta on B site. There is absolutely no description of an oxide composition in the '139 application that necessarily contains Fe or Co and at the same time Nb or Ta on the B site of the mixed conducting oxide in perovskite structure as claimed.

Application No.: 09/695,232

Docket No.: 21776-00055-US

The discovery of including small amounts of Nb and/or Ta in the oxide by applicants and the resulting stabilization of the perovskite type structure is not described in the '139 application. As applicants explain "[t]he inventors made the utmost effort to search a material of which a cubic crystal perovskite can exist constantly even if plenty of oxygen holes are introduced. As a result, the inventors found out that in a perovskite type crystal structure, a small amount of Nb and/or Ta ... positioned at the center of an oxygen octahedron (B site of the perovskite type structure) ... can be significantly stabilized. Page 26, beginning line 18.

Accordingly, applicants respectfully request that this rejection be withdrawn.

The '139 application also describes a method of producing the tubular solid-state membrane module. In particular, a method of sintering is described on page 13, lines 6 to 45 (EXAMPLE 3). The sintering of the dense mixed conducting oxide layer is described in Step 3 as "The body is then fired at still higher temperatures to completely sinter the vacuum infiltrated mixed conducting oxide layer to produce a dense mixed conducting oxide layer". In other words, this description discloses a technology of sintering the dense continuous layer at higher temperatures than those used for sintering the porous body portion.

In the preparation of the claimed mixed oxides, applicants do the exact opposite. For example, claim 10 of the present application provides that the "porous body portion is sintered at a temperature higher than the sintering temperature for a dense continuous layer". As a result, the sintering process and temperatures used and claimed by applicants is based on a technical idea completely different from that disclosed in the '139 application. One advantage provided by the sintering process of the present application is that the deterioration of the porosity of the porous body portion during sintering of the dense continuous layer is minimized, or can be avoided altogether. This stated advantage is not provided by the teachings of the '139 application. Accordingly, applicants respectfully request that the rejection of claims 10, 17, 19, 41 and 42 directed to the method of producing a composite material be withdrawn.

Applicants submit that with respect to claim 16, the composition range defined by formula (2) lies within the composition range of the mixed conducting oxide  $A_xA'y'A''x''ByB'y'B''y''O_{3-z}$  described in claim 11 of the '139 application. However, as already stated the described mixed oxide in the '139 application encompasses several thousands, if not tens of thousands, different compounds. There is no specific disclosure to a more limited sub-genus or

Application No.: 09/695,232

Docket No.: 21776-00055-US

to a specific mixed conducting oxide expressed by  $\text{AFexO}_{(3-\delta)}$ . See, *In re Petering*, 301 F.2d 676, (C.C.P.A. 1966)

Furthermore, there is no description in the '139 application of a "porous body portion comprising a mixed conducting oxide" in the range expressed by  $\text{AFexO}_{(3-\delta)}$  of formula (2). In the present application, applicants describe the difficulty of using a mixed conducting oxide in the composition range of formula (2) for a gastight dense continuous layer in a composite material for oxygen separation. However, the use of a mixed oxide of formula (2) exhibits excellent characteristic if it is used for a porous body portion of the oxide. See, page 33, line 13 through page 34 of the application. Accordingly, applicants respectfully request that the rejection of claims 16, 17, 19, 28-34, 39 and 40 be withdrawn.

The '139 application also describes providing a catalyzed layer on the surface of a dense mixed conducting oxide layer. Claim 16 and page 5, lines 18 to 26. Again, an extremely wide composition range is described for suitable catalysts - "metals and oxides of metals selected from Groups II, V, VI, VII, VIII, IX, X, XI, XV and the F Block lanthanides of the Periodic Table of the Elements according to the international Union of Pure and Applied Chemistry". However, there is no further limitation beyond this for the composition of the catalyzed layer.

In regard to claim 35 of the present application, the composition of the oxygen exchange layer is specifically limited to a composition which is expressed by a formula of  $\text{La}_u\text{Sr}_{b-u}\text{Fe}_v\text{Co}_{c-v}\text{O}_{3-w}$ , which differs from the composition of the mixed oxide, for oxygen isolation. Further, claim 39 of the present application provides an oxygen exchange layer which is limited to a composition that is different from that of the dense continuous layer on the surface of the dense continuous layer. Applicants go on to explain that by providing an oxygen exchange layer as claimed, a high oxygen permeation rate can be obtained. Applicants submit that it is impossible to deduce a preferable composition range for the oxygen exchange layer described in claims 35-37 and 39 of the present invention from the teachings of the '139 reference. Accordingly, applicants respectfully request that the rejection of claims 35-37 and 39 be withdrawn.

For the reasons described above, the teachings of the '139 application do not anticipate claims 1-8, 10-14, 16-17, 19, 20, 22-37 and 39-42. Accordingly, applicants respectfully request that the rejection be withdrawn.

Application No.: 09/695,232

Docket No.: 21776-00055-US

2. Cited Reference 2 [EP 0592809 A1]

EP 0592809 (the '809 application) discloses composite mixed conductor membranes for producing oxygen. The membranes include a dense layer and a porous layer. The porous layer of the membrane includes a multi-component metallic oxide whose composition is described in claims 4 to 7, and in the specification. Page 3, lines 43 to 51; page 5, lines 13 to 23; page 5, line 54 to page 6, line 4. The described composition range of this metallic oxide is  $A_xA'y'x'A''x''ByB'y'B''y''O_{3-z}$ .

Applicants submit that there is absolutely no teaching of a metal oxide composition which necessarily contain Fe or Co and at the same time Nb or Ta at the B site of the mixed conducting oxide in perovskite structure as claimed. Accordingly, applicants respectfully request that the rejection claims 1-8, 10-14, 20, 22-34, 39 and 40 be withdrawn.

Furthermore, the described metal oxide in the '809 application encompasses several thousands, if not tens of thousands, different compounds. The '809 application goes on to describe a preferred composition range of the oxide as  $A_xA'y'x'A''$ , which comprises a Group II metal selected from Ca, Sr, Ba and Mg. A more preferred composition is defined as  $La_xA_{1-x}Co_yFe_{1-y}O_{3-z}$ , and the most preferred compositions are defined as  $LaO \cdot 2BaO \cdot 8CoO \cdot 8FeO \cdot 2O_{3-x}$ ,  $PrO \cdot 2BaO \cdot 8CoO \cdot 8FeO \cdot 2O_{3-x}$ ,  $LaO \cdot 2BaO \cdot 8CoO \cdot 8CuO \cdot 2FeO \cdot 2O_{3-x}$ . Applicants submit that none of these more limited genres or species read on applicants' mixed oxide.

The '809 application also describes a method of depositing a dense layer on a porous substrate by sintering a slurry coating on the porous substrate. Page 7, lines 4 to 16. However, there is no description of using different sintering temperatures for the porous substrate and the dense layer. Claim 10 of the present application provides that the "porous body portion is sintered at a temperature higher than the sintering temperature for a dense continuous layer". Accordingly, applicants respectfully request that the rejection of claims 10, 17, 19, 41 and 42 be withdrawn.

Applicants submit that with respect to claim 16, the composition range defined by formula (2) lies within the composition range of the mixed conducting oxide  $A_xA'y'x'A''x''ByB'y'B''y''O_{3-z}$  described in claim 11 of the '809 application. However, as already stated the described mixed oxide in the '809 application encompasses several thousands, if not tens of thousands, different compounds. There is no specific disclosure to a more limited sub-genus or

Application No.: 09/695,232

Docket No.: 21776-00055-US

to a specific mixed conducting oxide expressed by  $\text{AFe}_x\text{O}_{(3-\delta)}$ . See, *In re Petering*, 301 F.2d 676, (C.C.P.A. 1966)

Furthermore, there is no description in the '139 application of a "porous body portion comprising a mixed conducting oxide" in the range expressed by  $\text{AFe}_x\text{O}_{(3-\delta)}$  of formula (2). In the present application, applicants describe the difficulty of using a mixed conducting oxide in the composition range of formula (2) for a gastight dense continuous layer in a composite material for oxygen separation. However, the use of a mixed oxide of formula (2) exhibits excellent characteristic if it is used for a porous body portion of the oxide. Such knowledge is not taught or suggested by the '809 reference. Accordingly, applicants respectfully request that the rejection of claims 16, 17, 19, 28-34, 39 and 40 be withdrawn.

The '809 application further describes a porous layer used to improve upon the oxygen flux of the membranes. The composition of the porous layer is described in claims 4 to 7, and in the specification. Page 3 lines 43 to 51; page 5 lines 13 to 23; and page 5, line 54 to page 6, line 4. In particular, line 54 on page 5 to line 4 on page 6, there is a description such that "The dense and porous layers of the composite membrane can be fabricated from the same or a different mixed conducting oxide"

In contrast, the composition of applicants' oxide exchange layer is defined as  $\text{La}_u\text{Sr}_{b-u}\text{Fe}_v\text{Co}_{c-v}\text{O}_{3+w}$ . See, claim 35. This claimed composition range provides a more effective porous layer, and is defined in relation to the composition of the dense layer such that "it is of different oxide composition than said oxide having oxide ion diffusivity (= dense layer)".

In claim 39 of the present application, the composition of oxygen exchange layer is further defined as "being made of an oxide of different composition than the oxide forming said dense continuous layer" in relation to the composition of the dense continuous layer. By defining the composition by the more narrow compositional range, applicants achieve a "higher oxygen permeation rate". Such knowledge is not taught or suggested in the '809 application. See, *In re Papesch*, 315 F.2d 381 (C.C.P.A. 1963)

Further, in claims 1 and 2, and from line 5 to line 14 on page 6 of the '809 application, there is a disclosure of morphology of the porous layer. The porous layer is described as having an "average pore radius is less than about 10 micrometers, the thickness of the porous layer is from 1 micrometer to about 2 millimeters. In contrast, the morphology of applicants' oxygen

Application No.: 09/695,232

Docket No.: 21776-00055-US

exchange layer is defined as having a "porosity is from 20% to 80%; thickness is 30 micrometers or less, preferably 10 micrometers or less". See, claims 35 and 36. Accordingly, applicants respectfully request that the rejection of claims 35 to 37 and 39 be withdrawn.

For the reasons described above, the teachings of the '809 application do not anticipate claims 1-8, 10-14, 16-17, 19, 20, 22-37 and 39-42. Accordingly, applicants respectfully request that the rejection be withdrawn.

### 3. Cited Reference 3 [WO 00/48717]

WO 00/48717 (the '717 application) discloses "a method of manufacturing a membrane which selectively transports oxygen". The compositions of the substrate and the film or coating are described in claim 7 and on page 4, lines 8 to 12 of the specification. Once again, the described composition range is quite broad,  $A_xA'y'A''x''ByB'y'B''y''O_{z+d}$ , and the described oxides encompass several thousands, if not tens of thousands, different compounds. Moreover, the only reference to a more limited material composition is that  $La_2NiO_4$ . See, Examples 1 to 3 on pages 7 to 8. The oxide,  $La_2NiO_4$ , is known as a mixed conducting oxide having a  $K_2NiF_4$ -type crystal structure.

Applicants claim an oxide of perovskite-type structure. Although these two mixed conducting oxides include similar elements, they are entirely different materials with different crystal structures. Accordingly, applicants respectfully request that the rejection of claims 1-8, 10-14, 20, 22-27, 29-34, 39 and 40 be withdrawn.

The '717 application also describes sintering a film on a porous substrate. Examples 3 to 5, pages 8 and 9. The production method as described includes "The coating was deposited by dipping the substrate into the slip. The coated substrate was dried, slowly heated to 500°C, thermal treated at 1100°C, and heated to 1300°C to densify the film." Consequently, the porous substrate and the film are subjected to the same heat treatment at the same time, and thus are sintered simultaneously.

Claim 10 of the present application requires that the "porous body portion is sintered at a temperature higher than the sintering temperature for a dense continuous layer". As a result, the sintering process and temperatures used and claimed by applicants is distinguishable from that

Application No.: 09/695,232

Docket No.: 21776-00055-US

disclosed in the '717 application. One advantage provided by the sintering process of the present application is that the deterioration of the porosity of the porous body portion during sintering of the dense continuous layer is minimized, or can be avoided altogether. This effect is not obtained from the teachings of the '717 application. Accordingly, applicants respectfully request that the rejection of claims 10, 17, 19, 41 and 42 directed to the method of producing a composite material be withdrawn.

Furthermore, the '717 application does not describe providing an oxygen exchange layer to improve the oxygen permeation rate. Accordingly, applicants respectfully request that the rejection of claims 35 to 37 and 39 be withdrawn.

For the reasons described above, the teachings of the '717 application do not anticipate claims 1-8, 10-14, 16-17, 19, 20, 22-37 and 39-42. Accordingly, applicants respectfully request that the rejection be withdrawn.

#### 4. Cited Reference 4 [U.S. 5,439,706]

U.S. 5,439,706 (the '706 patent) discloses "a method for manufacturing an inorganic membrane by Organometallic Chemical Vapor Deposition". The oxide composition of the material, which demonstrates oxide ion conductivity, is described in column 3, lines 1 to 31. The composition range is again quite broad,  $A_xA'_{1-x}B_yB'_{1-y}O_{3-z}$ . A preferred composition range is described as  $La_xA_{1-x}Co_yFe_{1-y}O_{3-z}$ . As in the other three cited references there is no description of the composition containing Fe or Co and at the same time containing Nb or Ta on the B site. Accordingly, applicants respectfully request that the rejection of claims 1 to 8, 23, 25 and 39 be withdrawn.

Furthermore, the described oxide,  $A_xA'_{1-x}B_yB'_{1-y}O_{3-z}$ , encompass several thousands, if not tens of thousands, different compounds, and there is no specific description of an oxide with the composition range of formula (2).

The '706 patent also describes heat treatment (sintering) conditions. Column 5, line 63 to column 6, line 27. However, there is no teaching or suggestion to use one heat treatment condition for the porous body and then another different heat treatment for the dense continuous layer. In particular, there is no teaching of heating conditions that includes "a porous body

Application No.: 09/695,232

Docket No.: 21776-00055-US

portion [that] is sintered at a temperature higher than the sintering temperature for [the] dense continuous layer", as embodied in claim 17 of the present application. Accordingly, applicants respectfully request that the rejection of claim 17 be withdrawn.

Furthermore, the '706 patent does not describe providing an oxygen exchange layer to improve the oxygen permeation rate. Accordingly, applicants respectfully request that the rejection of claims 35 to 37 and 39 be withdrawn.

For the reasons described above, the teachings of U.S. 5,439,706 do not anticipate claims 1-8, 10-14, 16-17, 19, 20, 22-37 and 39-42. Accordingly, applicants respectfully request that the rejection be withdrawn.

In view of the above, each of the presently pending claims in this application is believed to be in immediate condition for allowance. Accordingly, the Examiner is respectfully requested to withdraw the outstanding rejections of the claims and to pass this application to issue.

The Director is hereby authorized to charge any fees, or credit any overpayment, associated with this communication, including any extension fees, to CBLH Deposit Account No. 22-0185.

Date: August 20, 2004

Respectfully submitted,

By 

Joseph Barrera

Registration No.: 44,522

CONNOLLY BOVE LODGE &amp; HUTZ, LLP

1990 M Street, N.W., Suite 800

Washington, DC 20036-3425

(202) 331-7111

(202) 293-6229 (Fax)

Attorneys for Applicant